

(19) World Intellectual Property Organization  
International Bureau(43) International Publication Date  
17 January 2002 (17.01.2002)

PCT

(10) International Publication Number  
WO 02/05361 A1(51) International Patent Classification<sup>7</sup>: H01L 51/20,  
51/40, H05B 33/04(81) Designated States (national): AE, AG, AL, AM, AT, AT  
(utility model), AU, AZ, BA, BB, BG, BR, BY, BZ, CA,  
CH, CN, CR, CU, CZ, CZ (utility model), DE, DE (utility  
model), DK, DK (utility model), DM, DZ, EE, EE (utility  
model), ES, FI, FI (utility model), GB, GD, GE, GH, GM,  
HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KR (utility  
model), KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG,  
MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD,  
SE, SG, SI, SK, SK (utility model), SL, TJ, TM, TR, TT,  
TZ, UA, UG, UZ, VN, YU, ZA, ZW.(21) International Application Number: PCT/US00/31393  
(22) International Filing Date:  
15 November 2000 (15.11.2000)

(25) Filing Language: English

(26) Publication Language: English

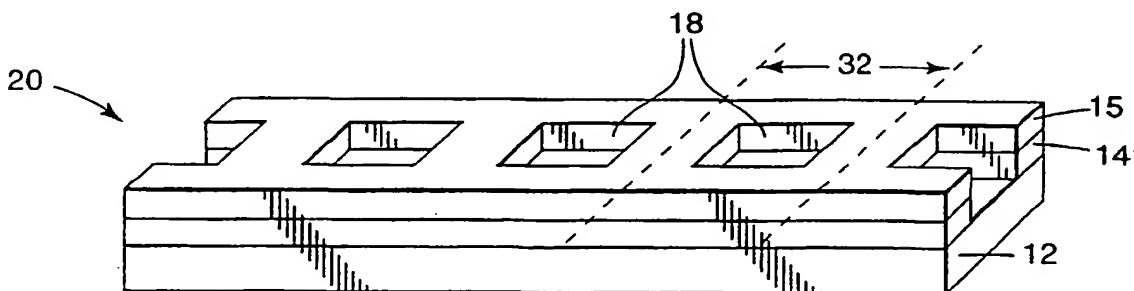
(30) Priority Data:  
09/614,993 12 July 2000 (12.07.2000) US

(71) Applicant: 3M INNOVATIVE PROPERTIES COMPANY [US/US]; 3M Center, Post Office Box 33427, Saint Paul, MN 55133-3427 (US).

(72) Inventors: MCCORMICK, Fred B.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). BAUDE, Paul E.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). VERNSTROM, George D.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

(74) Agents: GOVER, Melanie G. et al.; Office of Intellectual Property Counsel, P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

(54) Title: ENCAPSULATED ORGANIC ELECTRONIC DEVICES AND METHOD FOR MAKING SAME



A1

**(57) Abstract:** The invention provides methods for making encapsulated organic electronic devices (OED) including organic light emitting diodes (OLED). The present invention can provide a robust OED device by means of in situ edge sealing enhancing structural integrity and device lifetime. The edge sealing is provided by using an adhesive component (14) applied to a substrate (12) prior to OED element (16) deposition. In one OLED embodiment of the present invention, a thin layer of an adhesive (pressure sensitive adhesive, hot melt, or curable) is applied to release liner, openings are cut in the adhesive/liner composite, then the composite is adhered to an electrode-coated substrate. Alternatively, an adhesive may be applied directly onto the electrode-coated substrate, e.g., by printing in a desired pattern, optionally partially cured or dried, then covered with one or more liners that act as a mask during deposition of the OLED elements. Another method would be to prepare a blank liner with a patterned adhesive, then die cut openings complementary to the adhesive pattern in the liner to allow deposition of OLED elements once the adhesive/liner is placed on the substrate.

BEST AVAILABLE COPY

5

## ENCAPSULATED ORGANIC ELECTRONIC DEVICES AND METHOD FOR MAKING SAME

10

### TECHNICAL FIELD

This invention relates to a method for making an encapsulated organic electronic device.

15

### BACKGROUND

Organic electronic devices (OED)s are articles that include layers of organic materials, at least one of which can conduct an electric current. Illustrative examples of known OED constructions include photovoltaic devices, rectifiers, transmitters, and organic light emitting diodes (OLED)s.

An organic light emitting diode (OLED) is a typical example of an OED. OLEDs, sometimes referred to as lamps, are desirable for use in electronic media because of their thin profile, low weight, and low driving voltage, *i.e.*, less than about 20 volts. OLEDs have potential use in applications such as backlighting of graphics, pixelated displays, and large emissive graphics.

OLEDs typically consist of an organic light emitter layer and additional organic charge transport layers on both sides of the emitter, all of which are sandwiched between two electrodes: a cathode and an anode. The charge transport layers comprise an electron transporting layer and a hole transporting layer. Charge carriers, *i.e.*, electrons and holes, are injected into the electron and hole transporting layers from the cathode and anode, respectively. Electrons are negatively charged atomic particles and holes are vacant electron energy states that behave as though they are positively charged particles. The charge carriers migrate to the emitter layer, where they combine to emit light.

Examples of OLEDs include molecularly doped polymer devices where charge carrying and/or emitting species are dispersed in a polymer matrix (*see* J. Kido, "Organic Electroluminescent devices Based on Polymeric Materials," *Trends in Polymer Science*, 1994, 2, 350-355), conjugated polymer devices where layers of polymers such as

poly(phenylenevinylene) act as the charge carrying and emitting species (*see* J. J. M. Halls, D. R. Baigent, F. Cacialli, N. C. Greenham, R. H. Friend, S. C. Moratti, and A. B. Holmes, "Light-emitting and Photoconductive Diodes Fabricated with Conjugated Polymers," *Thin Solid Films*, **1996**, *276*, 13-20), vapor deposited small molecule heterostructure devices (*see* U. S. Pat. No. 5,061,569, and C. H. Chen, J. Shi, and C. W. Tang, "Recent Developments in Molecular Organic Electroluminescent Materials," *Macromolecular Symposia*, **1997**, *125*, 1-48), light emitting electrochemical cells (*see* Q. Pei, Y. Yang, G. Yu, C. Zang, and A. J. Heeger, "Polymer Light-Emitting Electrochemical Cells: In Situ Formation of Light-Emitting p-n Junction," *Journal of the American Chemical Society*, **1996**, *118*, 3922-3929), vertically stacked organic light-emitting diodes capable of emitting light of multiple wavelengths (*see* U. S. Pat. No. 5,707,745, and Z. Shen, P. E. Burrows, V. Bulovic, S. R. Forrest, and M. E. Thompson, "Three-Color, Tunable, Organic Light-Emitting Devices," *Science*, **1997**, *276*, 2009-2011).

Essentially all organic light emitting materials, organic charge transport materials and organic hole transport materials are adversely affected by heat, light, oxygen, and moisture. The low work function metal cathodes typically used in OLEDs are also sensitive to oxygen and moisture, which can cause corrosion and failure of the cathode. It is important, therefore, to protect these layers from exposure to the open air. Some methods of making OEDs such as OLEDs partially protect these layers, for example, methods that use barrier films as substrates, but the top layer of the OLED remains exposed. A separate encapsulation step, such as bonding a metal cap on top of an OLED, is typically required. This separate encapsulation step adds to manufacturing complexity and is not generally suited to the fabrication of flexible OLEDs.

25

## SUMMARY OF INVENTION

The present invention describes a novel protected OED and a novel method of making a protected OED. The method of the present invention protects the applied layers of an OED from exposure to the open air, particularly during manufacture of an OLED, in order to assure the maximum possible lifetime of the device.

30

In one aspect, the present invention provides a method for making an organic electronic device comprising:

providing an adhesive-coated release liner from which a pattern has been cut,

applying the adhesive-coated side of the patterned release liner to an electrode substrate to form a composite structure having at least a portion of the electrode substrate exposed,

depositing one or more organic electronic elements on the exposed electrode of the composite structure,

removing the release liner from the composite structure, and

adhering a sealing layer to the exposed adhesive of the composite structure.

In another aspect, the invention provides a method for making an organic electronic device comprising;

coating an adhesive in a pre-determined pattern on an electrode substrate to form a composite structure wherein areas of the substrate remain exposed;

optionally at least partially curing, or drying, the adhesive;

applying a liner mask to the patterned adhesive such that at least a portion of the exposed substrate areas remain exposed;

depositing one or more organic electronic elements on the masked composite structure;

removing the liner mask from the adhesive; and

applying a sealing layer.

In yet another aspect, the invention provides an article comprising an organic electronic device wherein the layers between the anode and cathode are surrounded by an adhesive layer and wherein the circumference of the adhesive layer is equal to the circumference of one or both of the electrode substrate or sealing layer.

In another aspect, the present invention provides a method for making an OED such as an OLED wherein an encapsulating component, which is incorporated into the OED structure, also serves as a mask while the OED is being constructed. Pursuant to one aspect of the present invention, an adhesive-coated liner, having openings cut through it in the desired shape of the OED, is applied, adhesive side down, to the OED substrate.

In another aspect of the invention, the adhesive may be precisely applied to the OED substrate in a liquid form, e.g., by an inkjet printing method or screen printing. One or more solid liner layers, having openings complementary to the openings in the adhesive layer may then be applied. After this step, the desired element layers of the OED are

sequentially deposited onto the liner/adhesive/substrate composite structure. Once all the desired OED layers have been applied, the liner is removed from the adhesive, leaving complete or substantially complete OED structures surrounded by an adhesive structure.

In yet another aspect of the invention, an OED may be an OLED. The present 5 invention will allow an OLED to be constructed from the anode or cathode side. In either case, a sandwich structure can be made by applying the adhesive layer to an electrode layer then depositing the OLED elements in the order necessary to construct an OLED.

In a further aspect, the OLED may be partially constructed on each electrode substrate and the final operational OLED obtained by laminating the two partial lamps 10 together. The sandwich structure would be attained by first applying the adhesive/liner layer on one or both electrode substrates and depositing, for example, hole transport material onto the anode substrate and electron transport and emitter material on the cathode substrate and subsequently aligning and laminating said films together with heat and/or pressure. The final OLED device can have enhanced structural stability over other 15 types of laminated OLED devices due to the adhesive forming the *in situ* edge seal.

In some embodiments, it may be preferable to have the organic OLED elements coated over an area larger than the underlying electrode layer. Having the organic layers cover a larger area than the electrodes helps avoid electrical shorting.

After the OLED organic layers have been applied, a counter electrode is applied. 20 If the counter electrode is a cathode, the electrode may comprise a sheet of electrically conductive material, *e.g.*, aluminum, that is placed on the adhesive structure to form a sealing layer, thereby encapsulating each OLED device. If the counter electrode is an anode, the electrode may comprise a sheet of electrically conductive material, *e.g.*, ITO on a polymer film substrate, that is placed on the adhesive structure to form a sealing layer, 25 thereby encapsulating each OLED device. If the sealing layer is a counter electrode, it must touch the entire top organic element layer to provide illumination of the total area of the OLED device. In these cases, the counter electrode will also act as a barrier to water and air. Alternatively, the counter electrode may be deposited within the boundaries of the adhesive structure and a protective material may be placed on top of the adhesive 30 structure, thereby encapsulating each OLED device. The protective layer may be conducting or non-conducting. If it is conducting, it may be used as a conducting lead, and it must touch at least one point of the adjacent electrode layer. The resulting sheet of

OLED devices may then be cut in patterns defined by the adhesive portion, thereby providing individual OLED devices, each of which is encapsulated between two substrates that are edge sealed by an adhesive.

5 As used in this invention:

“partially cure” means increasing the viscosity to induce film formation or reduce creep or flow, *i.e.*, wherein the physical state of the composition is altered, such as by transforming from a fluid to less fluid state, going from a tacky to a non-tacky state, going from a soluble to insoluble state, or decreasing the amount of polymerizable material by its consumption in a chemical reaction.

An advantage of at least one embodiment of the present invention is that it allows an OED to be made without exposure to the atmosphere at any time during the process, for example by conducting the deposition steps in a vacuum.

15 Another advantage of at least one embodiment of the present invention is that it allows for encapsulated OEDs to be made on a continuous basis.

Another advantage of at least one embodiment of the present invention is that it provides an OED having structural stability.

20 Another advantage of at least one embodiment of the present invention is that because the OED is not exposed to the atmosphere while being constructed, air and water sensitive materials can be used in the OED.

Another advantage of at least one embodiment of the present invention is that OEDs can easily be made in any desired shape.

25 Another advantage of at least one embodiment of the present invention is that OEDs can be made continuously in a roll-to-roll process.

Another advantage of at least one embodiment of the present invention is that OEDs may be made on a flexible substrate as compared to conventional methods that require a rigid substrate.

Although the invention described herein may be used for a wide variety of OED 30 devices, for illustrative purposes, only OLEDs will be discussed in detail.

Other features and advantages of the invention will be apparent from the following drawings, detailed description, and claims.

## BRIEF DESCRIPTION OF DRAWINGS

Fig. 1 illustrates the substrate with the applied adhesive-coated liner mask.

Fig. 2 illustrates the final sheet of OLED devices after the OLED elements have  
5 been deposited, the liner has been removed, and the sealing layer has been applied to the exposed transfer adhesive. Lines where cuts can be made to convert the sheet of OLED devices into individual devices (*i.e.*, singulate) are also shown.

Fig. 3 illustrates an exploded view of the components of an organic electronic device made by a method of the present invention.

10 Fig. 4 illustrates a simplified representation of a vapor coating process that could be used in the present invention.

Fig. 5 illustrates a typical OLED construction without the encapsulating material of the present invention.

15 Fig. 6 illustrates a cross-sectional view of an OLED construction made using the present invention.

Fig. 7 illustrates a top view of the OLED of Fig. 6.

Fig. 8 illustrates a cross-sectional view of another OLED construction made using the present invention.

18 Fig. 9 illustrates a top view of the OLED of Fig. 8.

20 Fig. 10 illustrates the use of multiple adhesive/liner pairs of different sizes and shapes in OED fabrication.

Fig. 11 illustrates the use of adhesive and liner layers of different sizes in conjunction with directional layer deposition to control the area on which various deposited layers are applied.

25 Fig. 12 illustrates an OLED constructed by laminating together two partial OLED structures, at least one of which is made by a method of the present invention.

The present invention is susceptible to various modifications and alternative forms. Some specifics thereof have been shown by way of example in the drawings and will be described in detail. It should be understood, however, that the intention is not to limit the invention to the particular embodiments described. On the contrary, the intention is to cover all modifications, equivalents, and alternatives falling within the spirit and scope of

the invention as described by the following detailed description and as defined by the appended claims.

## DETAILED DESCRIPTION

5       The present invention can provide a robust OED device by means of *in situ* edge sealing thereby enhancing structural integrity and device lifetime. The edge sealing is provided by using an adhesive component applied to a substrate prior to OED element deposition.

10      In one OLED embodiment of the present invention, a thin layer of an adhesive (pressure sensitive adhesive, hot melt, or curable) is applied to a release liner, openings are cut in the adhesive/liner composite, then the composite is adhered to an electrode-coated substrate. Alternatively, an adhesive may be applied directly onto the electrode-coated substrate, *e.g.*, by printing in a desired pattern, optionally partially cured or dried, then covered with one or more liners that act as a mask during deposition of the OLED 15 elements. The OLED elements may be applied in one or more patterns (circles, rectangles, letters, numbers, bar code and so forth) through openings in the liner and the adhesive. If multiple masks having different sizes or shapes are used, the OLED element layers may be applied in different or overlapping areas of the opening, depending on when each mask is removed during the deposition process. Alternatively, the elements may be deposited 20 through an additional mask placed over the openings, which results in thin film deposition in a different, typically smaller, pattern than that defined by the adhesive liner mask described above. Another method would be to prepare a blank liner with a patterned adhesive, then die cut openings complementary to the adhesive pattern in the liner to allow deposition of OLED elements once the adhesive/liner is placed on the substrate.

25      Examples of suitable adhesives include pressure sensitive adhesives (PSA) made from acrylates such as Ultra-Clean Laminating Adhesive 501FL and Optically Clear Laminating Adhesive 8141 both available from 3M Bonding Systems Division, St. Paul, MN, rubbers such as KRATON styrenic block copolymers from Shell Chemicals, Houston, TX, silicones such as RHODOTAK 343 from Rhodia Silicones, Lyon, France, 30 and polyolefins such as poly(1-hexene), poly(1-octene), and poly(4-ethyl-1-octene) described in US 5,112,882; hot melt adhesives such as unfilled versions of the tackified polyamide-polyether copolymers described in U.S. Pat. No. 5,672,400 and the

thermoplastic polymer adhesive films described in U.S. Pat. No. 5,061,549; curable adhesives, thermosets, and crosslinking systems such as the unfilled versions of the epoxy/thermoplastic blends described in U.S. Pat. No. 5,362,421; the cyanate ester/ethylenically unsaturated semi-IPNs described in U.S. Pat. No. 5,744,557; the 5 epoxy/acrylate compositions described in WO 97/43352; two-part epoxies like ARALDITE 2011 from Ciba Specialty Chemicals, Brewster, NY; and UV curing resins like the Series 3100 adhesives from Three Bond of America, Inc., Torrance, CA. Various combinations of pressure sensitive adhesive, hot melt, and curable adhesives may be useful in the practice of the invention.

10 The adhesives may be filled with various particulates and fillers to provide special functionality such as controlling the bond line thickness, providing electrical or thermal conductivity, or desiccating the interior of the packaged OLED device. Filled adhesives would include thermally conductive adhesive such as 9882 Thermally-Conductive Adhesive Transfer Tape available from 3M Bonding Systems Division; electrically 15 conductive adhesives such as 9703 Conductive Adhesive Transfer Tape, 5303R Z-Axis Adhesive Film, and 7303 Z-Axis Adhesive Film, all of which are available from 3M Bonding Systems Division; and desiccant filled adhesives such as DESIMAX SLF Hot Melt films available from Multisorb Technologies Inc., Buffalo, NY. Filled adhesives may be used in conjunction with unfilled adhesives.

20 The adhesive may be applied to a release liner by any suitable coating method such as knife coating, extrusion, offset coating, spray coating, and curtain coating. The thickness of the adhesive will depend on the desired thickness of the finished product. Typical thicknesses for the adhesive structures range from about 500 to 0.05 micrometers. Thicknesses above or below this range may also be used as desired to suit the application.

25 Suitable release liner materials include polypropylene and fluoropolymer films, polymer films, such as polyester, coated with siloxanes or fluorocarbons, paper coated with siloxanes or fluorocarbons, metal foils coated with siloxanes or fluorocarbons, and metallized polymer films, such as metallized polyester, coated with siloxanes or fluorocarbons. The choice of liner will be dependent on its compatibility with subsequent 30 processing steps.

Patterns may be cut through the adhesive-coated liner by die cutting, laser cutting or other precision cutting methods such as kiss cutting. The patterns may be any desired

shape. More than one shape may be cut into a single sheet of adhesive-coated liner. Additionally, the shapes and sizes of material removed from the adhesive and liner need not be the same. For example, a square area may be removed from the adhesive layer while a smaller star-shaped area of the liner, located within the square area of the removed adhesive, may be removed from the liner layer such that after the OLED layers have been deposited through the star-shaped opening in the liner, a gap exists between the finished star-shaped OLED and the surrounding adhesive structure.

As shown in Figure 10, multiple layers of adhesives and liners may be used where each adhesive/liner layer has differently shaped openings to allow depositions in different areas at different stages of the fabrication process. Adhesive/liner mask layers **14a**, **15a** closest to the deposition substrate will typically have a larger opening than adhesive/liner mask layers **14b**, **15b** placed on top of layers **14a**, **15a**. In Figure 10, the organics for an OLED would be deposited onto ITO anode **46** with both adhesive/liner masks in place. Adhesive/liner mask layers **14b**, **15b** would then be removed and the cathode would be deposited through adhesive/liner mask layers **14a**, **15a**. Liner **15a** would be removed and replaced with a protective layer. The cathode would thus contact the ITO cathode pad **48** without any electrical shorts to the ITO anode **46**. Attaching the anode lead **46** and cathode lead **48** to a power supply would allow the final encapsulated OLED to operate. Adhesive/liner mask layers **14b**, **15b** could be removed after depositing one set of organic layers and be replaced by different adhesive/liner mask layers that had a different size or shape mask opening than adhesive/liner mask layers **14b**, **15b**. This would allow for more complex deposition patterns to be obtained, such as those required for multi-color devices. This could be particularly effective in a roll-to-roll process.

The patterned adhesive-coated liner may be applied by any convenient method, e.g., lamination methods such as hot, cold, pressure, and vacuum lamination, to a substrate thereby forming a mask on the substrate. Figure 1 illustrates a substrate **12** having a patterned transfer adhesive **14** with liner mask **15**. Transfer adhesive **14** and liner mask **15** both have openings **18** that extend through to substrate **12**. Cut lines **32** exemplify the location at which the finished sheet of devices will be singulated.

An alternative method of applying an adhesive layer to the substrate is to deposit a liquid adhesive by precision methods such as ink jet or screen printing in a desired pattern thereby leaving areas of substrate exposed, optionally partially curing or drying the

adhesive to reduce flow and prevent adhesive migration, then covering the adhesive with a solid film liner layer that has complementary patterned openings. By this method, the openings in the liner mask may be made bigger or smaller than the underlying exposed substrate area. The ability to make the liner mask and OLED structure different sizes can  
5 be advantageous. For example, to prevent electrical shorting it may be advantageous to make the interior OLED elements cover a larger area than the bottom and/or top electrode so the electrodes do not touch. In other instances, it may be preferable to make the mask openings smaller than the electrodes.

Figure 11 shows another construction method to avoid electrical shorting in an  
10 OLED. The method comprises evaporating, for example, organic layers 16, from direction A, and cathode layers 17, from direction B, onto substrate 12 which is coated with ITO anode 46 and ITO cathode pad 48. The shadowing effects of adhesive/liner mask 14/15 would allow for selective deposition of organic and cathode layers near the edges of the mask. This may be particularly effective when the opening in adhesive 14 is  
15 larger than the opening in liner 15.

Once adhesive/liner mask 14/15 is applied, one or more OLED elements may be deposited onto the masked substrate by any suitable coating method such as vacuum deposition, solution coating, and thermal transfer. These coating methods are further described in copending patent application USSN 09/389926, pp. 7-10. The layers of  
20 individual OLED elements may be from about 200 to about 5000 Å thick. In some cases, the thickness of the combined layers is approximately equal to the thickness of adhesive layer 14, for example when the final applied film is a metal foil that is a cathode or that makes electrical contact with the cathode layer. Having the thicknesses of the adhesive and OLED element layers be equal is also preferable when the adhesive does not flow  
25 much during bonding. This is typically true of pressure sensitive adhesives. However, when the adhesive flows well during bonding, as is typical with hot melt or curable adhesives, the thickness of the bond line, *i.e.*, the final thickness of the adhesive when the bond is complete, can be controlled by adjusting the curing chemistry and the bonding conditions, which control the flow of the adhesive, or by adding filler particles of a  
30 specific size, which thereby define the bond line thickness. Methods known in the art can be used to control the flow of the adhesive to obtain such adjustments. In such a case, the thickness of the adhesive may be greater than that of the OLED element layers.

A preferred encapsulated OLED structure may have conducting leads, such as indium tin oxide (ITO) leads extending under the bond line and making electrical contact with the anode and cathode as shown in Figures 6 and 7. In this case, the encapsulating film, *i.e.*, the final top layer, would not need to act as an electrical contact. In some 5 embodiments, leaving a gap between the protective layer and OLED, as is shown in Figures 6 and 7 may be desirable, for example when including a desiccant in the gap or when protecting the OLED from abrasion is desired. This may be achieved by having the adhesive layer substantially thicker than the OLED element layers.

The OLED element coating process may be conducted in a controlled atmosphere 10 such as nitrogen, argon, or vacuum. This conveniently allows OLEDs to be made with air- and water-sensitive materials such as the electroluminescent materials comprising OLEDs as described herein. The process allows the production of an encapsulated OLED that has not been contaminated by exposure to air. Subsequent lamination of the coated substrate to a sealing layer may provide a finished OLED that is edge-sealed and has 15 enhanced structural stability.

The suitability of a particular counter electrode, which may be used as a sealing 20 layer, will depend on the desired application and the other OLED elements. Examples of cathode counter electrode materials include calcium, lithium, aluminum, ITO, barium, ytterbium, samarium, magnesium/silver alloys, and multiple layer structures such as calcium/aluminum and copper phthalocyanine/ITO. Examples of anode counter electrode materials include ITO and gold. It is preferred that at least one counter electrode be transparent. Alternatively, the counter electrode may be coated onto the OLED construction as are the other OLED elements, after which a protective layer may be laminated over the counter electrode layer.

A sealing protective layer for an OLED may comprise, *e.g.*, dielectric/polymer and 25 metal/polymer layered composite films (sometimes referred to as polymer multilayer, or PML, films) such as those described in WO 00026973, US 5,725,909, US 4,954,371, and *Thin Solid Films*, 1997, 308-309 19-25, metal foils, metallized polymer films, dielectric coated polymer films such as SiO<sub>x</sub> coated polymer films (TECHBARRIER S available 30 from Mitsubishi Chemical Corporation, Tokyo), thin flexible glass such as 0.03 mm thick D 263 T glass available from Schott Displayglas GmbH, Mainz, Germany, and thin glass adhesively laminated to polymer films as described in WO 99/21708, metal plates, glass

plates, plastic plates, and fluoropolymer films such as ACLAR films available from Allied Signal Inc., Specialty Films, Pottsville, PA. Rigid protective layers, such as metal, glass, or plastic plates, may be more suited for batch OLED manufacturing processing. Flexible barrier films, such as thin glass, multilayer polymer films, such as those described in U.S. Pat. No. 4,954,371, metal foils, and metallized polymers may be preferred in many instances because they can be used in a roll-to-roll process as well as in a batch process. Depending on the intended use of the OLED, the protective layer need not necessarily be transparent or conducting.

Figure 2 illustrates an embodiment of an OLED construction after sealing layer 30 has been applied. Cut lines 32 are shown. Cut lines 32 indicate where the structure can be cut through the layers of sealing layer 30, transfer adhesive 14 and substrate 12 to provide individual OLEDs 10 such that the OLED elements 16 are encapsulated by adhesive 14 and sandwiched between substrate 12 and sealing layer 30.

An exploded view of an exemplary individual OLED structure made by the foregoing method is shown in Figure 3. Adhesive 14 on substrate 12 surrounds OLED elements 16. Sealing layer 30 forms the top layer of OLED construction 10. A conventional OLED construction without the encapsulating structure of the present invention is shown in Figure 5.

Suitable OLED substrates 100 include glass, transparent plastics such as polyolefins, polyethersulfones, polycarbonates, polyesters, polyarylates, and polymeric multilayer films, ITO coated barrier films such as the Plastic Film Conductor available from 3M Optical Systems Division, surface-treated films, and selected polyimides. It is highly desirable that the OLED substrate has barrier properties matching those of the protective (or counter electrode) film. Flexible rolls of glass may also be used. Such a material may be laminated to a polymer carrier for better structural integrity.

The anode material 102 coating the substrate is electrically conductive and may be optically transparent or semi-transparent. Suitable anode materials include indium oxide, ITO, fluorine tin oxide (FTO), zinc oxide, vanadium oxide, zinc-tin oxide, gold, platinum, palladium silver, other high work function metals, and combinations thereof.

The lamp elements that may be deposited on the masked anode-coated substrate include an optional hole injecting layer 104, a hole transporting layer 106, a light emitting

layer **108**, an electron transporting layer **110**, an optional electron injecting layer **112**, and a second electrode (cathode) **114**.

Suitable materials for the optional hole injecting layer **104**, which accepts holes from the anode and transmits them to hole transporting layer, include porphyrinic compounds, *e.g.*, copper phthalocyanine (CuPc), zinc phthalocyanine, acid doped poly(ethylenedioxythiophene) (PEDOT), and acid doped polyaniline.

Suitable materials for the hole transporting layer **106**, which facilitates the movement of holes from the anode layer **102** to the emitter layer **108**, include, *e.g.*, aromatic tertiary amine materials described in U.S. Pat. Nos. 5,374,489 and 5,756,224, such as 4,4',4''-tri(N-phenothiazinyl)triphenylamine (TPTTA), 4,4',4''-tri(N-phenoxazinyl)triphenylamine (TPOTA), N,N'-bis(3-methylphenyl)-N,N'-diphenyl benzidine (TPD), and polyvinyl carbazole.

Suitable materials for the emitter layer **108**, in which the holes and electrons combine to produce light, include, *e.g.*, tris(8-hydroxyquinolinato)aluminum (AlQ). The emission of light of different colors may be achieved by the use of different emitters and dopants in the emitter layer as described in the art (*see* C.H. Chen, J. Shi, and C.W. Tang "Recent Developments in Molecular Organic Electroluminescent Materials," *Macromolecular Symposia 1997 125*, 1-48).

Suitable materials for the electron transporting layer **110**, which facilitates the movement of electrons from the cathode to the emitter layer, include, *e.g.*, AlQ, bis(10-hydroxy-benzo(h)quinolinato) beryllium, bis(2-(2-hydroxy-phenyl)-benzolthiazolato) zinc, 3,4,5-triphenyl-1,2,4-triazole, 2-(4-biphenylyl)-5-(4-*t*-butylphenyl)-1,3,4-oxadiazole, and combinations thereof.

Suitable materials for the optional electron injecting layer **112**, which accept electrons from the cathode and transmit them to the electron transporting layer, include metal fluorides such as LiF, CsF, as well as SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, copper phthalocyanine (CuPc), and alkaline metal compounds comprising at least one of Li, Rb, Cs, Na, and K such as alkaline metal oxides, alkaline metal salts, *e.g.*, Li<sub>2</sub>O, Cs<sub>2</sub>O, and LiAlO<sub>2</sub>.

Suitable materials for the cathode **114**, which provides electrons, include, *e.g.*, LiF, Mg, Ca, Ag, Al, Li, Ba, Yb, Sm, and other low work function metals, alloys of Ca and Mg, ITO, FTO, zinc oxide, and lithium,

Alternatively, one or more layers of light emitting polymers such as poly(phenylenevinylene)s (PPV), polyfluorenes, and other light emitting polymers known in the art may replace layers 106, 108, and 110.

Once all of the desired OLED elements have been deposited, the liner may be removed to provide a substrate with an OLED construction surrounded by adhesive. Subsequent covering or sealing of this coated substrate with a protective layer would provide an OLED that is edge-sealed and has enhanced structural (and environmental) stability. Alternatively, the cathode layer may comprise a sheet of material that is applied to the adhesive matrix after the preceding OLED element layers have been deposited and the release liner has been removed. If a sheet of cathode layer material is used, this cathode layer can also serve as a sealing layer that provides edge sealing of the OLED structure. In such a case, no additional protective layer would be necessary.

The process of the present invention could be readily carried out using a roll-to-roll continuous web process as illustrated by Figure 4. For example, to prepare an OLED, a roll of flexible substrate 12 with pre-cut adhesive-coated liner mask 14/15 could be fed from roll 230 into vacuum chamber 210 of coater 200 where OLED elements could be deposited by vaporization from deposition sources 220 and 222, which may deposit the first two organic layers of an OLED, corresponding, for example, to layers 104 and 106 of the device shown in Figure 5. Alternatively, deposition source could furnish ITO anode layer 102 of Figure 5. This step would be followed by a second set of vapor depositions in chamber 212 from deposition sources 224 and 226. Typically, deposition sources 220 to 226 will supply all of the layers necessary to produce a working OLED device on substrate 12. It will be obvious to one of skill in the art that the number of deposition sources in processing line 200 may vary according to the OLED device to be built. This step will be followed removal of release liner 15 onto take-up roll 232 in take-up chamber 214 and application of sealing layer 30 from roll 234 onto exposed adhesive mask 14 to encapsulate and protect the OLED device. Sealing layer 30 may be a counter electrode or a protective layer, depending on the particular construction of the OLED device. Take-up chamber 214 can be under vacuum or can be at atmospheric pressure and filled with an inert atmosphere, such as argon or nitrogen. It will be obvious that the number of vacuum and take-up chambers may vary according to the desired set of process conditions. The finished sheet of OLED devices is then collected on take-up roll 236 from which it can be

further dispensed for conversion into individual OLEDs by singulation, as shown in Figures 1-3. Other arrangements and combinations of applied layers are also possible and would be apparent to one skilled in the art. The entire process could be carried out in a controlled atmosphere, such as under a vacuum or nitrogen, to minimize the possibility of 5 environmental damage to the OLED. The finished sheet of OLEDs could be removed from the coater and converted to individual OLEDs by cutting through the adhesively-bonded areas of the electrode and substrate films to give the final (or at least partially) packaged OLED of the general structure shown in Figure 3. Further encapsulation may also be done after fabrication. For example, after an electrode or protective layer is 10 laminated to the OLED structure, an additional protective encapsulating layer may be added by, for example, dipping the entire assembly into a liquid epoxy resin and curing said resin by heat and/or light.

The roll-to-roll process may be a solution process, a thermal transfer process, a 15 vacuum process, or various combinations thereof. For example, a conducting polymer buffer layer, such as PEDOT, could be solution coated (knife coat, curtain coat, micro gravure coat, screen printed, etc.) onto an ITO deposition substrate 12 with the adhesive/liner mask 14/15 in place. After drying, the organic OLED layers and the cathode layers could be deposited by vacuum deposition in chambers 210 and 212 onto the moving web. Liner mask 15 would then be removed to take-up reel 232 and in its place 20 protective layer 30 would be laminated to the newly formed OLED film to produce a roll of encapsulated OLED devices ready for singulation. Alternatively, OLED layers may be solution coated (knife coat, curtain coat, micro gravure coat, screen printed, *etc.*) onto an ITO deposition substrate with the adhesive/liner mask in place. After drying, the cathode layers may be deposited by vacuum deposition onto the moving web. The liner mask 25 could be removed to a take-up reel and in its place a protective layer would be laminated to the newly formed OLED film to produce a roll of encapsulated OLED devices ready for singulation.

The roll-to-roll process may be done on a continuously moving web or it may be accomplished by a step-and-repeat roll-to-roll process. A step and repeat process could 30 allow for more precise thickness control during the various vacuum deposition steps and could enable the use of more complex and finer featured shadow masks during the vacuum deposition steps. Different shadow masks could be easily moved in and out of position

during the deposition of the various layers while still maintaining the convenience of a roll-to-roll process. Step-and-repeat web processing will likely be slower than continuous web processing. A web with sprocket holes along the edges may be advantageous in step-and-repeat and continuous web processing.

5       The use of the in situ edge seal adhesives of the current invention can provide structural stability to OLEDs prepared by physical lamination of two substrates carrying the appropriate active layers. Although there are several ways to apply the current invention to the fabrication of OLEDs by lamination, only one will be exemplified herein. Such fabrication is illustrated by Figure 12. For structure 300, adhesive/liner mask  
10      314/315 may be placed on the metallized side of a polyester film 310 coated with aluminum 312 and placed into a vacuum deposition chamber. Approximately 5,000 Å of aluminum 316, 10 Å of LiF 318, 400 Å of AlQ 320, and 400 Å of 4,4'-bis(naphthalen-2-yl)-N-N-diphenyl benzidine ( $\alpha$ -NPD) 322 can be deposited in that order. Separately, for structure 400, polyester substrate 328 coated with ITO layer 326 can be coated with 1,000  
15      Å of aqueous poly(ethylenedioxythiophene) (PEDOT) solution 324 and allowed to dry. Both film structures may be transferred to an inert atmosphere glove box. Liner 315 may be removed from structure 300, which structure 300 may then be laminated by heat and pressure to structure 400. Completed OLED device 500 would thus be formed where an interface between active layers 322 and 324, in this example between a hole injecting layer  
20      and a hole transport layer, would be created by physical lamination of these two layers. The completed device has improved structural stability by virtue of the adhesive forming the in situ edge seal around the device.

Finely patterned and addressable devices may be fabricated by the methods of the current invention. For example, a passively addressable monochrome dot matrix display  
25      may be fabricated in a roll-to-roll process. ITO on polyester could be patterned by a combination of standard photolithography and acid etching into columns that run across the web. An adhesive/liner mask which contains rectangular openings that define the desired area of the display would be applied to the web. Small molecule OLED layers would be vacuum deposited onto the web or light emitting polymers solution coated onto  
30      the web to essentially cover the rectangular opening in the adhesive/liner mask. Cathode metal would then be vapor deposited through a slotted mask to create rows of cathode metal parallel to the web direction. The liner would then be removed and a protective film

laminated to the passive matrix display. Electrical connection to the anode would be simply made by contacting the columns of ITO that pass underneath the encapsulating adhesive. Electrical connection to the cathode rows could be made in a number of ways. The protective film laminated to the cathode side of the device could carry conductive traces that could be in registration with the rows of anode. These could be in physical contact with the cathode rows or could be attached by, for example, a Z-axis conductive adhesive film. Alternatively, the ITO substrate could be patterned to include ITO cathode contact pads, which pass underneath the encapsulating adhesive. The cathode rows can be contacted to these pads by, for example, the use of various deposition masks or by angle evaporation techniques.

This invention may also be useful to temporarily protect an OLED or a partial OLED device. Copending U.S. patent application 09/231723 describes the use of OLED donor sheets to create OLED devices by thermal transfers. These donor sheets may benefit from protection from the atmosphere to increase their shelf life. An adhesive/liner mask could be applied to a donor substrate prior to the deposition of the desired OLED layers. The liner could be removed and a protective film could be applied before the donor sheets are exposed to oxygen or moisture. The protective film could be removed from the donor film immediately prior to thermally imaging the donor sheet thereby minimizing or eliminating the exposure of the donor sheet to oxygen and moisture. It would be preferred that the donor substrate also have good oxygen and moisture barrier properties.

Another unforeseen advantage of the current invention is protection of the OLED and electrode layers from damage in a roll-to-roll process. The adhesive/liner mask will generally be thicker in the z direction than the various OLED and electrode layers. When the front face of the OLED film passes over various rollers in the web handling system required for a roll-to-roll process, the adhesive/liner mask can prevent or minimize the contact between the active OLED layers and these rollers. Furthermore, when the finished OLED film is wound up on the final take-up reel or "jumbo" roll, the adhesive/liner mask or the laminated protective film can prevent or minimize pressure induced damage to the OLED devices (or thermal donor films) as they are wound up on the final take-up reel.

It is also possible to establish electrical connections on the completed structure by removing some adhesive material from the outer edges of the OLED structure. When

anode and cathode leads are in the form of ITO or other conductive paths that pass under the adhesive bond line, electrical connections to drive the device are made by attaching the leads to a power supply or drive electronics. It may be desirable to remove portions of the protective layer and adhesive, which may cover these leads, to facilitate attachment of the  
5 power supply or drive electronics.

## EXAMPLES

Unless otherwise specified, all chemicals were obtained from Eastman Kodak,  
10 Rochester, NY.

### Example 1.

Figures 6 and 7 show an ITO-coated glass substrate, 5 cm x 7.6 cm x 1 mm (Thin Film Devices, Inc., Anaheim, CA) 42. The ITO coating 44 was divided into two strips of  
15 conductive areas 46 and 48 by removing a portion of the ITO coating approximately 10 mm wide across the width of the glass substrate by acid-etching. In addition, ITO was removed along the outer lengths of the substrate such that strip 46 was wider than strip 48, as illustrated in Figure 7. A portion of the substrate approximately 10 mm x 20 mm that spanned portions of both ITO strips 46 and 48 was isolated by hand-laminating onto the  
20 substrate a piece of pressure-sensitive adhesive transfer tape (3M™ Ultra Pure Viscoelastic Damping Polymer 242F01, 3M Company, St. Paul, MN) 50 having a release liner on its upper side (not shown) from which a central hole area had been removed, to form a shape like a picture frame. The upper release liner of adhesive 50 remained in place. Glass substrate 42 was rinsed with methanol and dried under a nitrogen flow, then placed in a spin-coater, covered with xylene, and spun dry. An aqueous solution of  
25 poly(ethylenedioxythiophene) (PEDOT) conductive polymer (BAYTRON, Bayer Corp., Pittsburgh, PA) 52 was applied to the cut-out area, spun-coated at 5000 rpm for 20 seconds, then dried at 100° C on a hotplate for 15 minutes. The coated slide was immediately placed in an evaporator vacuum chamber and the chamber was sealed and  
30 evacuated to approximately  $10^{-5}$  torr. A light-emitting construction was prepared on the PEDOT surface by vacuum deposition, in the following order, of: 100 Å layer of copper phthalocyanine (CuPc), at 1 Å/sec; 200 Å 4,4'-bis(naphthalen-2-yl)-N,N'- diphenyl

benzidine ( $\alpha$ -NPD) hole transport layer, at 3 Å/sec; 350 Å tris(8-hydroxyquinolinato)aluminum (AlQ) doped at approximately 1% during the first 200 Å with a coumarin dye (C545T<sup>TM</sup>) electron transport layer, at 2 Å/sec; 10 Å lithium fluoride, at 1 Å/sec; and 4000 Å aluminum, at 10 Å/sec, thereby creating organic thin films layer 54 and cathode layer 56.

In the vacuum chamber, organic layers 54 (CuPc,  $\alpha$ -NPD, and AlQ and coumarin dye) were coated directionally from one end of the chamber and a LiF/aluminum cathode 56 from the opposite end of the chamber. Deposition of organic layers 54 was shaded on one end of the coated substrate by the sample holder (not shown), leaving portions of strips 46 and 48 uncoated. Deposition of LiF/aluminum cathode 56 from the opposite end of the chamber allowed some of the LiF and aluminum to be deposited on the uncoated portion of the ITO layer 48, providing an electrical contact.

The device was removed from the vacuum chamber and placed in a glove box under nitrogen. The upper liner was removed from the adhesive 'frame' 50 that surrounded the deposited lamp, and a clean glass cover slide 58 was pressed in place on the exposed adhesive, completely covering the lamp while leaving ITO strips 46 and 48 exposed on each end of the substrate. The covered device was removed from the glove box and tested by connecting an electrical power source to ITO strips 46 and 48. Light was emitted from the lamp when current was passed through the contacts.

20

#### Example 2.

Figures 8 and 9 show an ITO-coated glass substrate, 5 cm x 7.6 cm x 1 mm (Thin Film Devices, Inc., Anaheim, CA) 72. Using standard electron beam evaporation methods, a layer of Al<sub>2</sub>O<sub>3</sub> approximately 1000 Å  $\mu$ m thick 74 was deposited on the ITO 76 using a shadow masking method that left an approximately 10 mm x 20 mm area of uncovered ITO 76a in the middle of substrate 72 and approximately 10 mm uncovered at one end of the substrate.

An electroluminescent lamp was constructed on the substrate as described in Example 1, wherein a portion of the Al<sub>2</sub>O<sub>3</sub> surrounding the central ITO area was surrounded by a pressure-sensitive adhesive transfer tape 78, and a lamp was prepared in the interior isolated area by depositing PEDOT layer 86 and organic films layer 80 and

aluminum anode layer 82. In the nitrogen glove box, the lamp was sealed by placing a sample of clean aluminum foil 84 on the exposed adhesive and in contact with the upper aluminum layer 82 of the lamp.

The device was removed from the glove box and tested by connecting an electrical power source to the aluminum foil encapsulating layer 82 (as the cathode) and to the uncovered ITO strip 76b at the end of the substrate (as the anode). Light was emitted from the lamp when current was passed through the contacts.

In the foregoing description, certain terms have been used for brevity, clarity, and understanding. No unnecessary limitations are to be implied therefrom beyond the requirement of the prior art because such terms are used for descriptive purpose and are intended to be broadly construed. Moreover, the description and illustration of the invention is by way of example, and the scope of the invention is not limited to the exact details shown or described.

What is claimed is:

1. A method for making an organic electronic device comprising:
  - a. providing an adhesive-coated release liner from which a pattern has been  
5 cut,
  - b. applying the adhesive-coated side of the patterned release liner to an electrode substrate to form a composite structure having at least a portion of the electrode substrate exposed,
  - c. depositing one or more organic electronic elements on the exposed electrode of the composite structure,
  - 10 d. removing the release liner from the composite structure, and
  - e. adhering a sealing layer to the exposed adhesive of the composite structure.
- 15 2. A method for making an organic electronic device comprising:
  - a. coating an adhesive in a pre-determined pattern on an electrode substrate to form a composite structure wherein areas of the substrate remain exposed;
  - b. applying a mask or release liner to the patterned adhesive such that at least a portion of the exposed substrate areas remain exposed;
  - 20 c. depositing one or more organic electronic elements on the composite structure;
  - d. removing the mask or release liner from the adhesive; and
  - e. applying a sealing layer.
- 25 3. The method of claim 1 or 2 wherein a protective film is applied to the composite structure after the release liner is removed, which protective film is removed prior to adhering of the sealing layer.
4. The method of claim 1 or 2 wherein the adhesive-coated liner is thicker in  
30 the z direction than the organic electronic elements.

5. The method of claim 1 or 2 wherein the adhesive is selected from the group comprising hot melt adhesives, pressure sensitive adhesives, curable adhesives, and filled adhesives.

5 6. The method of claim 5 wherein the filled adhesive is selected from the group comprising electrically conductive adhesives, thermally conductive adhesives, and desiccating adhesives.

10 7. The method of claim 6 wherein the filled adhesive is used in conjunction with an unfilled adhesive.

15 8. The method of claim 1 or 2 wherein the adhesive-coated liner is selected from the group comprising polypropylene films, metal foils coated with siloxanes or fluorocarbons, polyester coated with siloxanes or fluorocarbons, and fluoropolymer films.

9. The method of claim 1 or 2 wherein multiple adhesive-coated liners are applied to the electrode substrate and are removed at different stages during organic electronic element deposition.

20 10. The method of claim 2 wherein the adhesive is one or both of partially cured and dried between a. and b.

11. The method of claim 1 or 2 wherein the sealing layer is a counter electrode.

25 12. The method of claim 11 wherein the material comprising the counter electrode is selected from the group comprising lithium fluoride, calcium, aluminum, barium, ytterbium, samarium, lithium, indium tin oxide, fluorine tin oxide, zinc oxide, magnesium, silver, gold, and alloys of calcium and magnesium.

30 13. The method of claim 1 or 2 wherein the sealing layer comprises a partial organic electronic device construction that is laminated to the composite structure.

14. The method of claim 1 or 2 wherein a counter electrode layer is applied after the final organic element layer and the sealing layer is a protective layer.

15. The method of claim 14 wherein the adhesive-coated liner is thicker in the z direction than the organic electronic elements and the counter electrode layer.

16. The method of claim 14 wherein the material comprising the protective layer is selected from the group comprising metallized polymer films, polymer multilayer films, metal plates, foils, and thin flexible glasses.

10

17. The method of claim 15 wherein the material comprising the counter electrode is selected from the group comprising lithium fluoride, calcium, aluminum, barium, ytterbium, samarium, lithium, indium tin oxide, fluorine tin oxide, zinc oxide, calcium, magnesium, silver, gold, and alloys of calcium and magnesium.

15

18. The method of claim 2 further comprising the step of fully curing the adhesive after the device has been constructed.

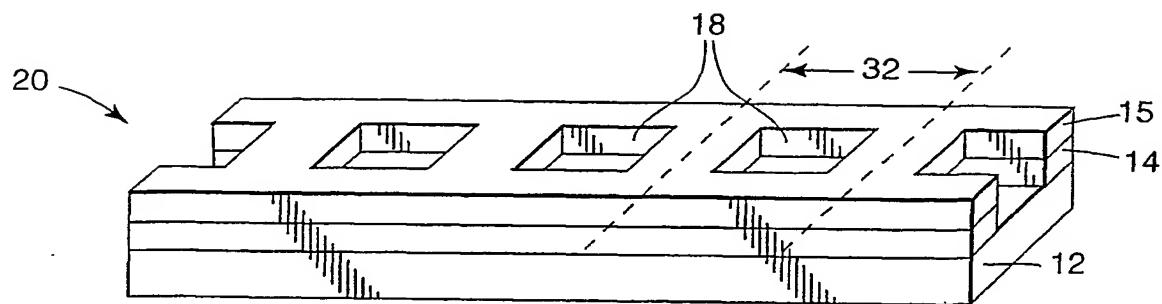
19. The method of claim 1 or 2 wherein the organic electronic device is an  
20 organic light emitting diode.

20. The method of claim 19 wherein the substrate is selected from glass and polymer multilayer films, the anode comprises indium tin oxide, the hole transporting layer comprises 4,4'-bis(naphthalen-2-yl)-N,N'-diphenyl benzidine, the light emitting  
25 layer comprises coumarin-doped tris(8-hydroxyquinolinato)aluminum, the electron transporting layer comprises, and the cathode comprises lithium fluoride and aluminum.

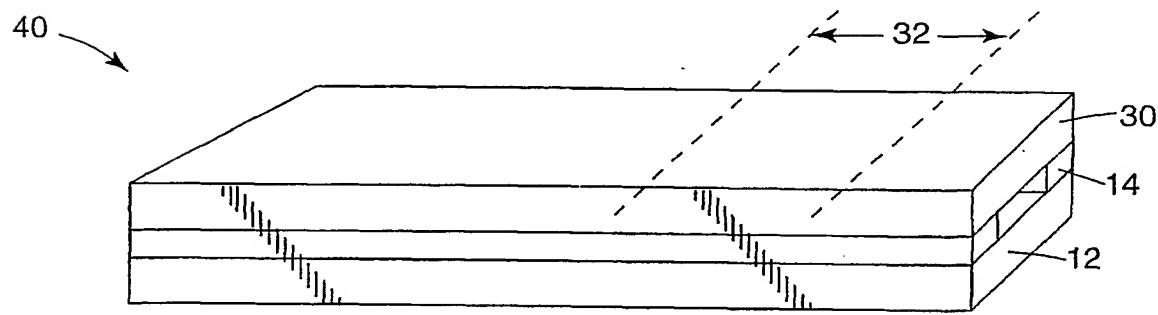
21. An article comprising an organic electronic device wherein the layers between the anode and cathode are surrounded by an adhesive layer and wherein the circumference of the adhesive layer is equal to the circumference of one or both of the  
30 electrode substrate or sealing layer.

22. The article of claim 21 wherein the circumference of the adhesive layer is greater than that of an electrode.

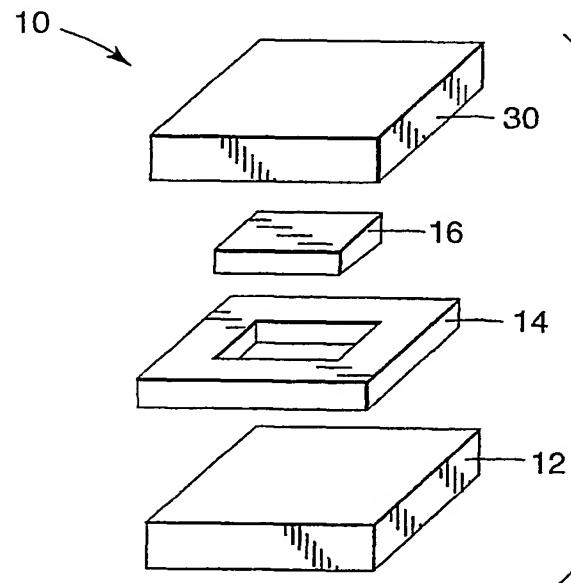
1/5



*Fig. 1*



*Fig. 2*



*Fig. 3*

2/5

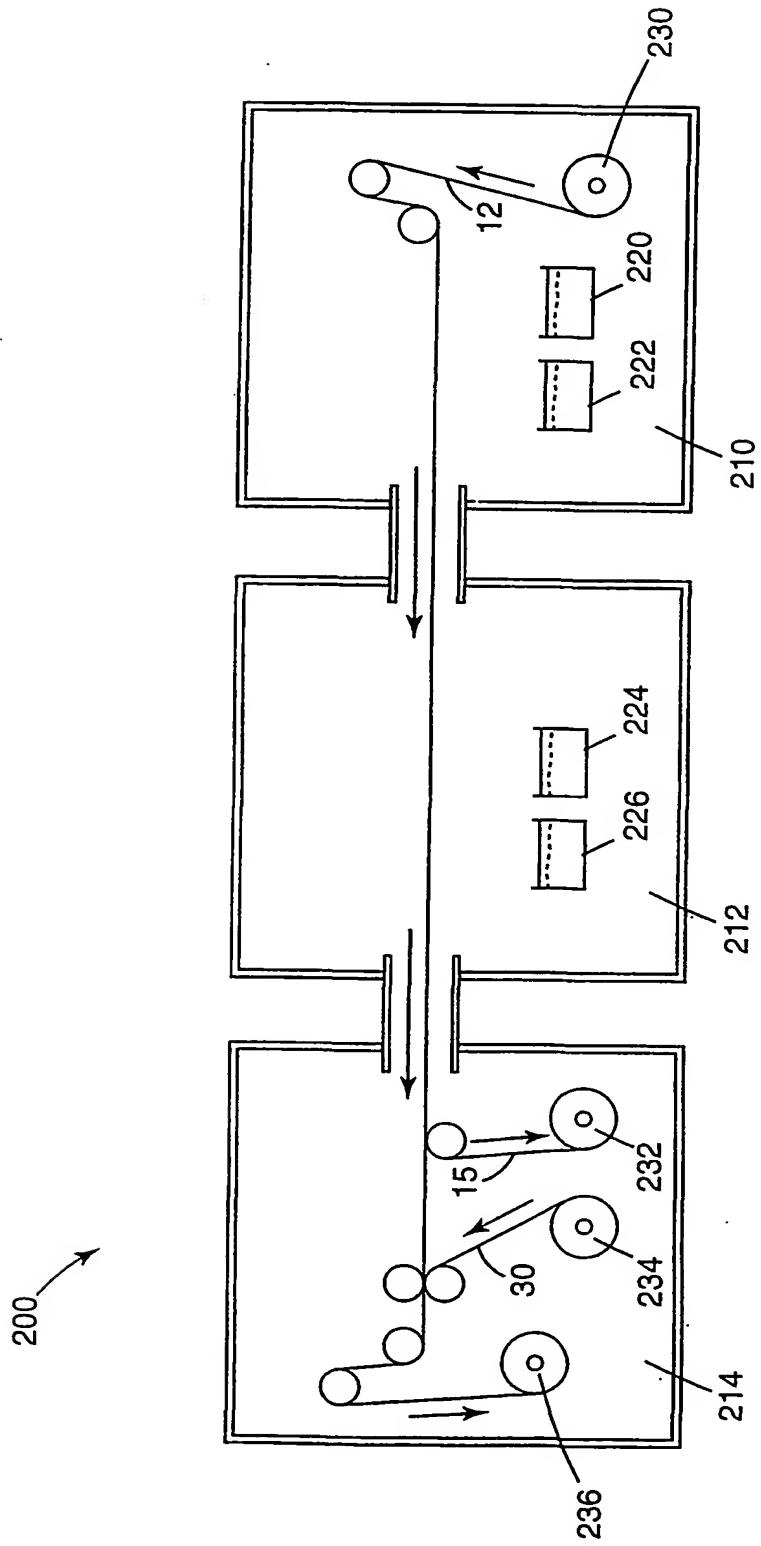
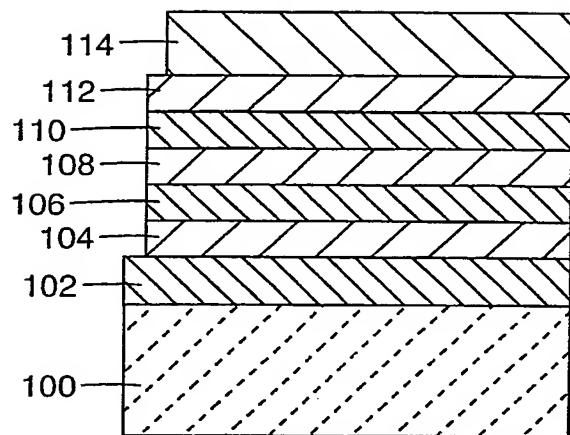
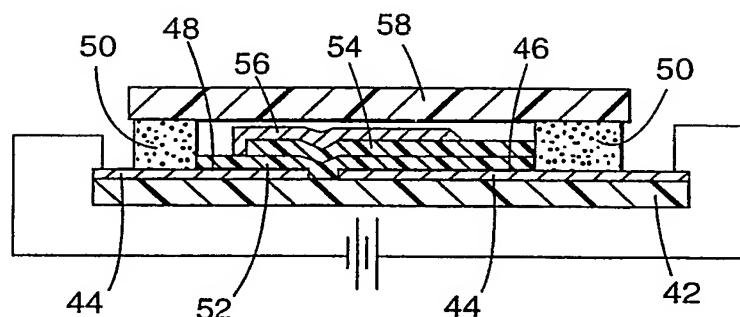
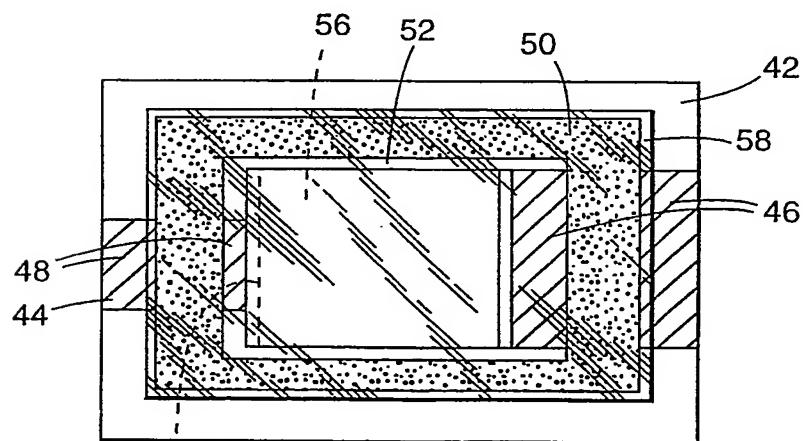
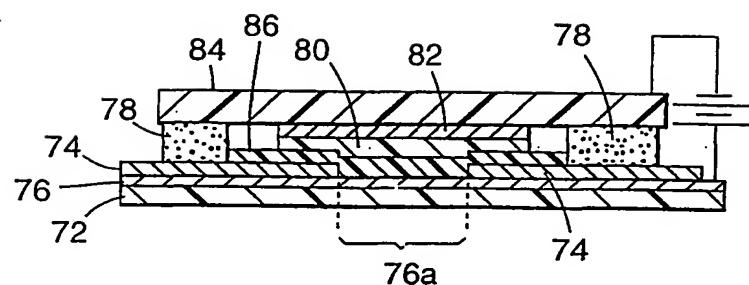


Fig. 4

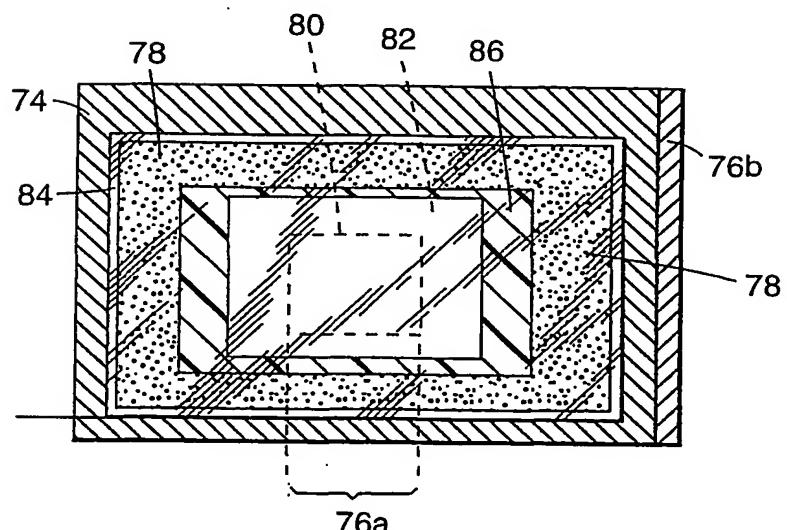
3/5

*Fig. 5**Fig. 6**Fig. 7*

4/5



*Fig. 8*



*Fig. 9*

5/5

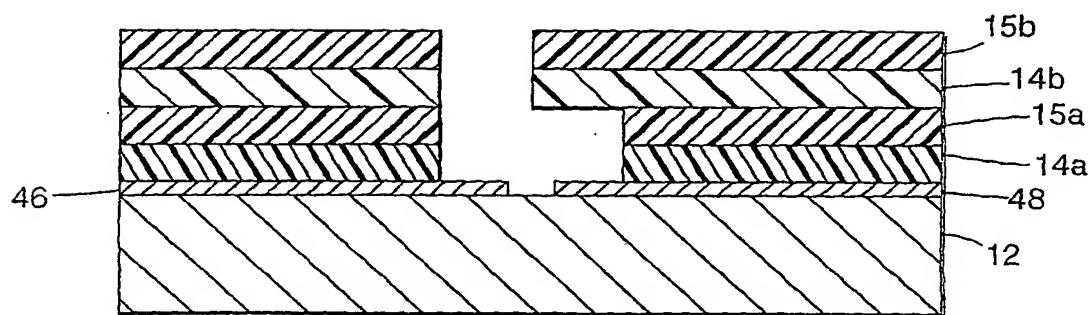


Fig. 10

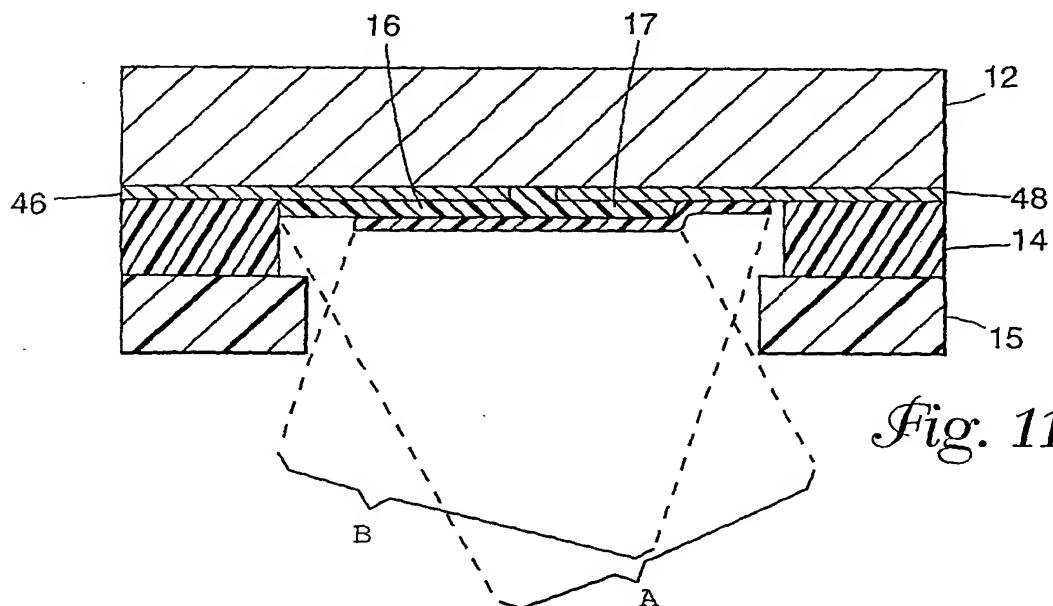


Fig. 11

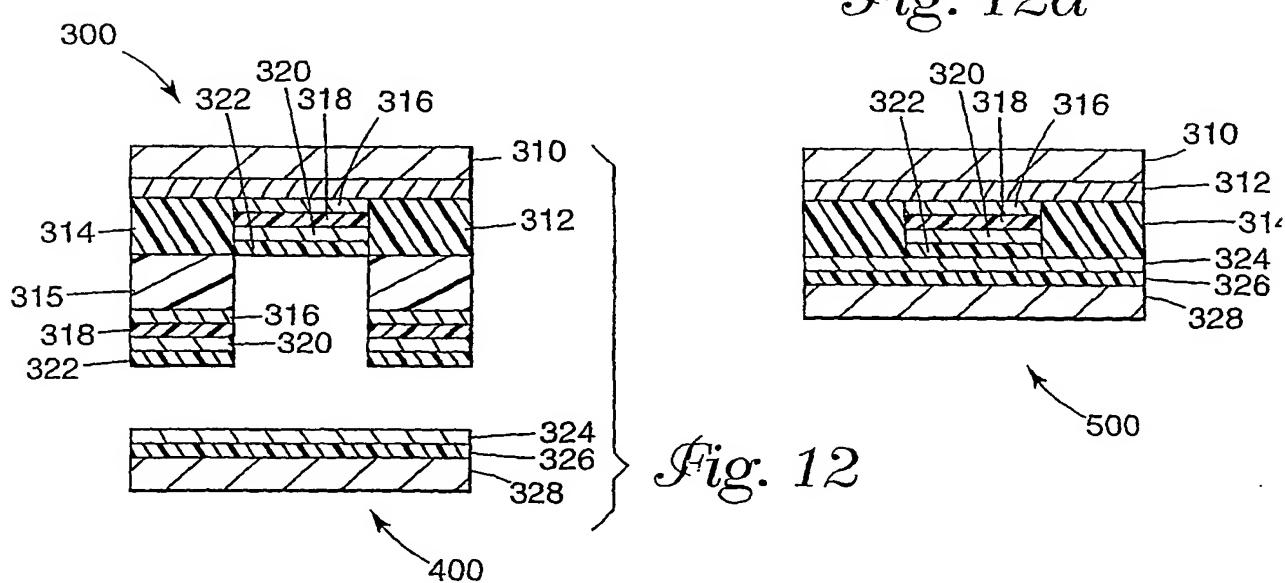


Fig. 12

## INTERNATIONAL SEARCH REPORT

Inter nal Application No  
PCT/US 00/31393A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 H01L51/20 H01L51/40 H05B33/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 H01L H05B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 786 664 A (LIU YOUNG) 28 July 1998 (1998-07-28) column 3, line 12 -column 4, line 60 ---	21
A	EP 0 884 930 A (IDEMITSU KOSAN CO) 16 December 1998 (1998-12-16)  the whole document ---	1, 2, 4, 5, 11-17, 19-21
A	EP 0 762 806 A (MOTOROLA INC) 12 March 1997 (1997-03-12)  the whole document ---	2, 4, 10, 11, 13-17, 19, 20 -/-

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*&\* document member of the same patent family

Date of the actual completion of the international search

31 May 2001

Date of mailing of the international search report

07/06/2001

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

van der Linden, J.E.

## INTERNATIONAL SEARCH REPORT

Inter nal Application No  
PCT/US 00/31393

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 00 05929 A (SEIKO EPSON CORP) 3 February 2000 (2000-02-03) -& EP 1 018 857 A (SEIKO EPSON CORP) 12 July 2000 (2000-07-12) the whole document ----	1,2,21, 22
P,X	EP 1 021 070 A (TDK CORP) 19 July 2000 (2000-07-19) the whole document -----	21,22

## INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 00/31393

Patent document cited in search report	Publication date	Patent family member(s)			Publication date
US 5786664	A 28-07-1998	NONE			
EP 0884930	A 16-12-1998	US 6175186 B	16-01-2001	WO 9731508 A	28-08-1997
EP 0762806	A 12-03-1997	US 5641611 A	24-06-1997	JP 9274452 A	21-10-1997
WO 0005929	A 03-02-2000	JP 2000100572 A	07-04-2000	CN 1286891 T	07-03-2001
		EP 1018857 A	12-07-2000		
EP 1021070	A 19-07-2000	JP 2000208252 A	28-07-2000		

BLANK PAGE

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- BLACK BORDERS**
- IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- FADED TEXT OR DRAWING**
- BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- SKEWED/SLANTED IMAGES**
- COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- GRAY SCALE DOCUMENTS**
- LINES OR MARKS ON ORIGINAL DOCUMENT**
- REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- OTHER:** \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**

**BLANK PAGE**